Measurement of Acidic Aerosol Species in Eastern Europe: Implications for Air Pollution Epidemiology

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A large number of studies have indicated associations between particulate air pollution and adverse health outcomes. Wintertime air pollution in particular has been associated with increased mortality. Identification of causal constituents of inhalable particulate matter has been elusive, although one candidate has been the acidity of the aerosol. Here we report measurements of acidic aerosol species made for approximately 1.5 years in Erfurt, Germany, and Sokolov, Czech Republic. In both locations, the burning of high-sulfur coal is the primary source of ambient air pollution. Twenty-four-hour average measurements were made for PM_{10},[particulate matter with an aerodynamic diameter (d_a) ≤10 µm], as well as fine particle (d_a <2.5 µm) H^+ and SO_4^{2-} for the entire study. Additionally, separate day and night measurements of fine particle H^+, SO_4^{2-}, NO_3^-, and NH_4^+ and the gases, SO_2, HNO_3, HONO, and NH_3 were collected with an annular denuder/filter pack system over a 7-month (late winter–summer) period with additional measurements during pollution episodes the following winter. At both sites, 24-hr SO_2 (mean concentrations of 52 µg/m^3, with peak levels of >585 µg/m^3) and PM_{10} (mean concentration 60 µg m^3) concentrations were quite high. However, aerosol SO_4^{2-} concentrations (mean concentration of approximately 10 µg/m^3) were not as great as expected given the high SO_2 concentrations, and acidity was very low (mean concentration of <1 µg/m^3, with peak levels of only 7 µg/m^3). Low acidity is likely to be the result of NH_3 neutralization and slow conversion of SO_2 to SO_4^{2-}. These data, along with evidence that aerosol acidity exposures are significantly lower than ambient levels and the reported association between fine particulate air pollution and health outcomes in regions where little aerosol acidity has been measured, suggest that particulate acidity alone is not the primary component defining fine particulate air pollution toxicity. *Key words*: acid aerosols, air pollution, Eastern Europe, epidemiology, particulates. *Environ Health Perspect* 103:482–488 (1995)

Beginnning more than 50 years ago, winter episodes of high levels of air pollution were associated with excess mortality in a number of locations, such as London; Donora, Pennsylvania; and the Meuse Valley in Belgium. Although comprehensive air quality data were often not available for these episodes, analysis suggested that although all fine particle concentrations were elevated, acidic aerosols in particular, in addition to sulfur dioxide, were the constituents associated with effects (*1,2*). The potential effect of acidic aerosol exposure has been investigated in North America, where in the summer acidic species are produced via photochemical oxidation mechanisms (*3*). One motivating factor for focusing on summer acid aerosol episodes in North America was the suggestion that the historical winter fog episodes produced high levels of acidic aerosols (*1,4*).

As a result of severe air pollution episodes, control strategies were implemented to reduce the burning of coal for residential heating in London, high-sulfur coal was replaced with low-sulfur varieties, and emission control strategies were implemented, such as the addition of tall stacks which emit above inversion layers. These strategies have greatly reduced the severity of localized wintertime episodes in western Europe and North America. In contrast, the use of tall stacks and the increased demand for summertime power (for air conditioning, for example) has led to long-range transport of sulfur oxides above inversion layers. As this is a summertime phenomenon, photochemical oxidation mechanisms are responsible for production of acidic aerosol species (*3*). Therefore, although the production mechanisms are different, the same agent that was suspected in earlier winter episodes has also been investigated for North American summer situations, even though typical summertime levels of aerosol acidity in North America are significantly lower than in the historical winter fog episodes.

Recently, several studies have attempted to examine the relative importance of aerosol acidity for the health effects of air pollution. Pope and colleagues have shown that wintertime air pollution, especially fine particulate matter ≤10 µm in diameter (PM_{10}, d_a ≤ 10 mm), is associated with excess mortality, as well as increased school absences and hospital admissions in an area of Utah (*5–8*). Although these studies showed elevated levels of wintertime air pollution, levels of acidic aerosols were extremely low (*7*). Dockery et al. examined the association among PM_{10}, aerosol acidity, and gaseous pollutants with daily mortality in eastern Tennessee and St. Louis, Missouri (*9*). The authors found associations of similar magnitude for PM_{10} in both locations and weaker associations with aerosol acidity (*9*). The consistency of the PM_{10} associations with daily mortality is striking given that mean aerosol acidity levels in eastern Tennessee were more than three times higher than the mean levels in St. Louis, although PM_{10} and aerosol sulfate concentrations were similar. It is also important to note that in eastern Tennessee and St. Louis, PM_{10} concentrations peak during the summer months, whereas in the Utah Valley, elevated PM_{10} is a winter phenomenon.

Dockery and colleagues also reported on the relationship between mortality and air pollution in six U.S. cities (*10*). In this analysis, mortality was more strongly associated with levels of fine and inhalable particulates, as well as sulfate particles, than it was with aerosol acidity (*10*). Somewhat different results were presented by Thurston et al. for analysis of Toronto, Ontario, data: aerosol acidity showed a stronger relationship to summer hospital admissions than did other particle measures such as aerosol SO_4^{2-}, PM_{10}, or $PM_{2.5}$ (*11*). Such results are perplexing, especially when one considers that even when aerosol acidity is measurable in the ambient air, exposure is likely to be low due to neutralization in indoor environments (*12,13*).

Another opportunity to investigate the impact of aerosol acidity and the epidemiological association between winter smog and health came with the end of Communist rule in Eastern Europe. A major air pollution epidemiology study was undertaken in former East Germany and in the Czech Republic. An initial analysis of data from the period 1980–1989, just before the reunification of Germany, has associated excess mortality in Erfurt, Germany, with levels of SO_2 and suspended particulates (*14*). Although air pollution data were available for these pollutants, there were no measurements available of fine particulates nor of the

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chemical composition (such as acidity) of the particulates. With the reunification of Germany in 1989 and the end of Communist rule in the neighboring Czech Republic, the opportunity arose not only to conduct a more detailed epidemiological study, but also to collect measurements of aerosol acidity and fine particulate matter (*15*). During 1990–1992, the effect of ambient SO_2 and total suspended particulates (TSP) on peak flow, medication intake, and daily respiratory symptoms in a population of 160 asthmatic children and 110 adults with asthma or chronic bronchitis was analyzed. A small but consistent decrease in evening peak flow and an increase in daily symptoms was associated with elevated levels of SO_2 and TSP, whereas the effects of aerosol acidity and SO_4^{2-} were generally weaker (*15*).

In the Czech Republic, several studies have indicated effects of ambient air pollution on health. Bobak and Leon report an ecological study in which associations were observed between PM_{10}, and to a lesser extent SO_2 and NO_x, and post-neonatal respiratory mortality in Czech infants (*16*). Lower pulmonary function and a higher prevalence of respiratory symptoms in 2nd-, 5th-, and 8th-grade children living in the highly polluted region of northern Bohemia were observed when compared to the lung function and respiratory symptoms of children living in the less polluted area of southern Bohemia (*17*). In northern Bohemia, high levels of PM_{10} and SO_2 (12-hr averages of 800 and 1100 $\mu g/m^3$) were measured in a 1993 winter episode. During this period SO_4^{2-} levels were also quite high (200 $\mu g/m^3$) and the fine aerosol was acidic, although only a few percent of the SO_4^{2-} was in the form of sulfuric acid (Stevens RK, personal communication).

Ambient air measurements of PM_{10}, acidic aerosols, acidic gases, and NH_3 were conducted for approximately 1.5 years in Erfurt, Germany, and Sokolov, Czech Republic. This study provided a unique opportunity to examine levels of acidic aerosol species in environments directly impacted by both local and regional sources of high sulfur (up to 5%) lignite "brown coal." Monitoring was specifically focused on winter inversions during which pollutant concentrations were expected to be highest. Here we report on these measurements and compare them to our substantial database of acid aerosol measurements in North America and the available data for Europe. We also compare characteristics of the chemical composition of these Eastern European winter atmospheres to other coal-burning regions and with summer episodes in North America. Our purpose was to determine if aerosol acidity can be measured in wintertime air pollution episodes in locations where the burning of high-sulfur coal is a major air pollution source. Our primary hypothesis was that the health effects associated with wintertime air pollution in similar settings are associated with acidic aerosols. Extensive measurements of acidic aerosol species were collected to support an indirect test of this hypothesis.

Methods

Two cities were selected for air monitoring, Erfurt, in the former East Germany, and Sokolov, in the Czech Republic. Both Erfurt and Sokolov were reported to be subject to winter inversions, which resulted in poor ambient air quality (high TSP and SO_2 concentrations) and reduced visibility. Erfurt (population approximately 220,000) is northwest of Frankfurt, approximately 100 km east of the former east–west border, and is a regional center of commerce. The city has an older, central area where the primary heating source is individual coal furnaces. The outer areas of the city contain large apartment complexes with steam heat supplied by a large coal-burning power plant located several kilometers east of the city center. Erfurt is situated on a flat plain bordered by a 100-m high ridge on all sides but north. The sampling site was 1 km from the town center, 15 m from the nearest structure, and 30 m from the nearest major road. Sampler inlets were located approximately 2 m above ground level.

Sokolov (population approximately 60,000) is an industrial town in a coal-mining region of the Czech Republic, 100 km east of the German–Czech border, at the southern edge of the northern Bohemia region where some of the largest SO_2 sources in Europe are located. In the immediate vicinity of Sokolov are several power plants and large industrial complexes in the region, in addition to a coal gasification plant. Sokolov is in the broad Ohre river valley, which is bordered by 400-m high valley walls. The sampling site was located on the terrace of a two-story building approximately 2 km from the central district. Sampler inlets were located 2 m above the terrace surface.

Particulate acidity (measured in the $PM_{2.5}$ particulate size fraction) and PM_{10} samples were collected with the Harvard Impactor (with the addition of a citric acid-coated honeycomb denuder for acidity sampling) (*18,19*). The Harvard-EPA Annular Denuder System (HEADS) was used to measure gaseous and particle species during some portions of the study. Sampling and analysis procedures are reported in detail elsewhere (*20,21*). Daily or every second day 24-hr samples of fine particulate (d_a <2.5 μm) mass, H^+, and SO_4^{2-} were collected from December 1990–June 1992, and 24-hr PM_{10} samples were collected February 1991–June 1992. Annular denuder measurements were made twice daily (0800 hr–1600 hr, 1600 hr–0800 hr) from February 1991 to September 1991 and during episode periods from October 1991 to April 1992. Detection limits for 12-hr denuder measurements were 16.2 $\mu g/m^3$, 1.2 $\mu g/m/^3$, 0.8 $\mu g/m^3$ (as H_2SO_4), 2.3 $\mu g/m^3$, 0.6 $\mu g/m^3$ and 0.3 $\mu g/m^3$, for SO_2, NH_3, H^+, SO_4^{2-}, NH_4^+, and NO_3^-, respectively. The detection limit for PM_{10} was 9 $\mu g/m^3$, and the Harvard Impactor 24-hr detection limits were 0.4 (as H_2SO_4) and 1.4 $\mu g/m^3$ for H^+ and SO_4^{2-}, respectively.

One of the features of the annular denuder system is a multistage filter pack which includes a Teflon filter to collect the fine particles as well as backup Na_2CO_3 and citric acid-coated filters to collect gaseous HNO_3 and NH3, respectively, that have evolved from the collected particles as the result of artifact reactions (*22*). Backup filters are used to determine whether any aerosol acidity has been "lost" due to particle–particle interactions. For example, acidic sulfate aerosols that react with NH_4NO_3 aerosols will be neutralized, and gaseous HNO_3 will be liberated (Eq. 1). An additional source of volatilized HNO_3 is from the dissociation of particulate NH_4NO_3 into gaseous HNO_3 and NH_3 (Eq. 2).

$$H_2SO_4\,(p), NH_4HSO_4\,(p), (NH_4)_3H(SO_4)_2\,(p) + (NH_4)NO_3\,(p) \rightarrow (NH_4)_2SO_4\,(p) + HNO_3\,(g) \quad (1)$$

$$NH_4NO_3\,(p) \rightarrow NH_3\,(g) + HNO_3\,(g) \quad (2)$$

By quantifying the amount of HNO_3 (as NO_3^-) and NH_3 (as NH_4^+) on the backup filters, the measured acidity can be corrected for any acidity that is lost due to particle–particle neutralization reactions.

Results and Discussion

Tables 1–3 present summary statistics and compare the winter and summer concentrations measured in Erfurt and Sokolov to those measured in 23 North American communities as part of a major epidemiological study (*23*). These sites were used for comparison because the studies used the same or equivalent measurement and analytical techniques for aerosol acidity as the Eastern European sites, measurements were collected over an entire year, and all measurements were of 24-hr duration. Additional measurements of aerosol acidity in North America have been reported previously (*3*) and demonstrate that the 23 North American communities used for comparison represent a reasonable description of the range of aerosol acidity and

Table 1. Summer (May–September) and winter (November–March) concentrations of particulate matter ≤10 μm (PM_{10}; μg/m^3) measured in Sokolov, Erfurt, and 23 communities in the United States and Canada[a]

	Summer						Winter				
Site	N	Mean	SD	Min	Max	Site	N	Mean	SD	Min	Max
Penn Hills, PA	73	46.3	24.6	6.4	119.3	**Erfurt**	**76**	**63.5**	**42.9**	**12.2**	**208.9**
Erfurt	**53**	**44.3**	**19.6**	**16.5**	**89.3**	**Sokolov**	**72**	**54.2**	**34.7**	**3.2**	**171.2**
Sokolov	**36**	**40.9**	**19.7**	**9.2**	**84.1**	Monterey, CA	72	32.6	11.2	13.7	61.1
Simi Valley, CA	71	39.8	11.7	15.2	73.5	Livermore, CA	74	30.6	17.2	3.1	83.8
Morehead, KY	71	39.5	17.9	8.6	77.1	Parson, WV	73	29.6	17.7	5.2	93.6
Zanesville, OH	65	39.2	17.1	10.9	84.9	Simi Valley, CA	74	28.9	14.4	6.8	64.6
Springdale, AZ	65	37.3	13	0	67.9	Hendersonville, TN	68	27.9	12.7	8.9	60.2
Hendersonville, TN	75	37.1	15.7	10.6	77.3	Uniontown, PA	72	27.8	11.5	10.9	70.5
Uniontown, PA	76	36.5	19.3	14.5	116.5	Penn Hill, PA	61	26.4	11.9	4.9	76.7
Athens, OH	76	34.8	16.4	10.7	99.5	Springdale, AZ	71	26	15	0	89.3
Blacksburg, VA	73	30.9	11.4	5	60.5	Morehead, KY	68	24.9	9.2	8.5	49.5
Parson, WV	76	30.5	14.5	8.1	73.3	Leamington, ON	70	23.4	11.5	7	78.5
Dunville, ON	68	30.3	18.8	0	83.3	Newtown, CT	59	23.3	11.9	0	58.5
Oak Ridge, TN	64	29.6	11.3	13.3	64.4	Athens, OH	75	23	9.4	5.2	49.6
Leamington, ON	77	28.6	13.1	9.5	78.8	Blacksburg, VA	70	22.7	8.7	5.5	41.9
Charlottesville, VA	74	27.2	12.6	7	61.3	Oak Ridge, TN	65	21.8	10	3.6	53.7
Newton, CT	73	27.1	17.7	0	79.4	Zanesville, OH	66	21	10.3	1.7	58.1
State College, PA	73	26.8	13.1	0	67.3	Penticton, BC	72	19.9	9.9	6.5	47.6
Livermore, CA	70	26.3	10.9	4.8	53.2	Charlottesville, VA	62	19.8	8.2	7.5	60.1
Aberdeen, SD	78	25	11.4	5.3	74.2	Dunville, ON	58	17.1	9.5	0	47.7
Egbert, ON	77	23.2	13.5	0	70.8	Egbert, ON	68	16.7	9.4	4.6	51.3
Yorktown, SK	73	22.8	9.7	5.1	48.6	State College, PA	72	16.6	6.8	0	38.6
Pembroke, ON	75	21.2	14.2	0	64	Aberdeen, SD	67	16.5	8.4	3.4	41.2
Monterey, CA	71	20.3	9.8	0	45.5	Pembroke, ON	47	16.1	11.4	0	51.1
Penticton, BC	73	18.9	9.1	4	47	Yorktown, SK	60	14.1	6.1	1.7	34

[a]For each season, the sampling sites are listed in order of highest mean concentration.

Table 2. Summer (May–September) and winter (November–March) concentrations of aerosol strong acidity (μg/m^3 as H_2SO_4) measured in Sokolov, Erfurt, and 23 communities in the United States and Canada[a]

	Summer						Winter				
Site	N	Mean	SD	Min	Max	Site	N	Mean	SD	Min	Max
Uniontown, PA	69.0	4.3	5.3	0.1	39.1	Oak Ridge, TN	65.0	1.3	1.5	0.0	8.8
Parson, WV	71.0	4.0	4.7	0.0	19.2	Uniontown, PA	71.0	1.2	1.1	0.2	6.5
Morehead, KY	72.0	3.8	3.3	0.1	14.8	Parson, WV	68.0	1.1	1.0	0.1	4.5
Athens, OH	75.0	3.8	4.0	0.1	24.6	Hendersonville, TN	71.0	1.1	1.2	0.0	7.2
Penn Hills, PA	66.0	3.4	4.2	0.0	19.4	Morehead, KY	69.0	1.1	1.0	0.2	5.3
Oak Ridge, TN	70.0	3.3	2.9	0.0	14.3	State College, PA	71.0	1.0	0.9	0.0	4.1
State College, PA	66.0	3.3	3.2	0.4	16.5	Charlottesville, WV	68.0	0.9	0.9	0.0	4.5
Zanesville, OH	59.0	3.2	3.7	0.0	17.8	Zanesville, OH	70.0	0.8	1.0	0.0	6.7
Hendersonville, TN	70.0	3.2	2.5	0.2	10.9	Athens, OH	75.0	0.7	0.7	0.0	4.2
Charlottesville, VA	66.0	3.1	2.7	0.0	15.5	Pembroke, ON	43.0	0.6	0.5	0.0	1.7
Blacksburg, VA	72.0	2.9	2.2	0.3	10.9	Blacksburg, VA	71.0	0.6	0.6	0.0	2.8
Dunville, ON	57.0	2.6	3.4	0.0	13.9	Simi Vally, CA	70.0	0.6	0.9	0.0	5.6
Newtown, CT	70.0	2.0	2.2	0.0	9.1	Newton, CT	63.0	0.6	0.5	0.0	2.2
Leamington, ON	73.0	1.5	2.1	0.0	13.5	Livermore, CA	72.0	0.6	0.4	0.0	1.6
Pembroke, ON	64.0	1.4	2.2	0.0	13.3	Leamington, ON	73.0	0.5	0.6	0.0	3.0
Springdale, AR	71.0	1.0	0.6	0.0	2.6	Monterey, CA	72.0	0.5	0.3	0.1	1.5
Simi Valley, CA	68.0	0.9	0.6	0.0	3.0	Penn Hills, PA	63.0	0.5	0.5	0.0	2.4
Sokolov	**74.0**	**0.5**	**0.3**	**0.0**	**1.5**	Penticton, BC	70.0	0.4	0.3	0.0	1.4
Livermore, CA	73.0	0.5	0.4	0.0	1.9	Dunville, ON	54.0	0.4	0.4	0.0	2.3
Egbert, ON	48.0	0.5	1.2	-0.4[b]	6.0	**Sokolov**	**72.0**	**0.3**	**0.6**	**0.0**	**4.8**
Monterey, CA	72.0	0.4	0.3	0.0	1.5	Springdale, AZ	67.0	0.3	0.3	0.0	1.0
Penticton, BC	69.0	0.4	0.2	0.0	1.1	**Erfurt**	**74.0**	**0.2**	**0.3**	**0.0**	**1.7**
Erfurt	**75.0**	**0.4**	**0.3**	**0.0**	**1.4**	Aberdeen, SD	66.0	0.0	0.1	-0.3	0.4
Aberdeen, SD	74.0	0.2	0.2	-0.2	0.9	Yorktown, SK	30.0	0.0	0.1	-0.3	0.2
Yortown, SK	75.0	0.0	0.2	-1.2	0.5	Egbert, ON	16.0	-0.1	0.1	-0.4	0.1

[a]For each season, the sampling sites are listed in order of highest mean concentration.
[b]Negative values indicate alkaline samples.

aerosol SO_4^{2-} concentrations found in North America.

European measurements are extremely limited and more difficult to compare because of different sampling and analytical techniques and small sample sizes. Using similar techniques to those described here, Hoek et al. found low levels of aerosol acidity (0–8.8 μg/m^3 as H_2SO_4) and aerosol SO_4^{2-} (1–24 μg/m^3) in an extensive monitoring study conducted in The Netherlands (*24*), and Kitto and Harrison measured aerosol acidity levels of 0–8.7 and SO_4^{2-} concentrations of 1–48 μg/m^3 in a coastal area of southeast England (*25*). In a limited series of measurements, Puxbaum, et al. (*26*) measured 6- to 12-hr averages of wintertime aerosol acidity and SO_4^{2-} concentrations in Ljublijana (formerly Yugoslavia), in a rural site in Italy (Po Valley), and at a suburban

Table 3. Summer (May–September) and winter (November–March) concentrations of aerosol sulfate (μg/m^3) measured in Sokolov, Erfurt, and 23 communities in the United States and Canada[a]

	Summer						Winter				
Site	*N*	Mean	SD	Min	Max	Site	*N*	Mean	SD	Min	Max
Penn Hills, PA	62.0	12.1	9.2	0.0	42.6	**Erfurt**	**74.0**	**8.4**	**6.5**	**0.9**	**30.5**
Uniontown, PA	70.0	10.4	8.1	0.0	51.5	**Sokolov**	**72.0**	**7.7**	**5.4**	**1.4**	**23.8**
Morehead, KY	72.0	10.1	5.4	1.7	27.4	Athens, OH	74.0	4.6	2.4	0.4	12.0
Athens, OH	74.0	10.0	7.7	0.0	43.1	Uniontown, PA	71.0	4.4	2.2	1.2	11.9
Zanesville, OH	60.0	9.9	7.7	0.0	37.5	Oak Ridge, TN	66.0	4.2	2.4	0.9	12.9
Hendersonville, TN	69.0	9.6	5.1	2.4	22.5	Penn Hills, PA	63.0	4.1	2.1	0.0	10.7
Blacksburg, VA	72.0	9.4	5.3	1.2	23.8	State College, PA	71.0	4.0	2.4	0.5	10.0
Dunville, ON	59.0	9.1	7.9	0.0	30.6	Morehead, KY	69.0	3.8	2.2	1.1	9.7
State College, PA	67.0	9.1	7.0	0.6	29.5	Zanesville, OH	70.0	3.8	2.2	0.7	11.3
Parson, WV	71.0	8.6	7.7	0.0	33.8	Parson, WV	67.0	3.8	1.8	1.0	10.3
Oak Ridge, TN	70.0	8.6	5.7	0.0	29.4	Blacksburg, VA	70.0	3.6	1.7	0.6	9.9
Sokolov	**67.0**	**8.6**	**5.0**	**1.4**	**21.2**	Hendersonville, TN	73.0	3.5	2.4	0.0	13.2
Charlottesville, VA	67.0	8.2	5.4	0.0	26.4	Charlottesville, VA	68.0	3.5	1.9	0.7	9.2
Erfurt	**69.0**	**8.0**	**4.7**	**1.5**	**21.5**	Leamington, ON	73.0	3.4	1.9	0.0	8.7
Newtown, CT	69.0	6.3	6.0	0.0	26.0	Newtown, CT	61.0	3.3	2.0	0.0	9.7
Leamington, ON	74.0	6.3	5.9	0.0	33.1	Dunville, ON	53.0	3.3	1.7	0.0	8.2
Springdale, AR	72.0	4.8	2.9	0.0	12.8	Egbert, ON	16.0	2.4	1.5	0.6	6.8
Egbert, ON	48.0	4.7	7.2	0.0	30.7	Springdale, AR	66.0	2.4	1.6	0.0	8.2
Pembroke, ON	67.0	4.4	5.6	0.0	27.8	Simi Valley, CA	70.0	2.2	3.3	0.0	14.8
Simi Valley, CA	63.0	4.0	2.5	0.0	10.0	Pembroke, ON	41.0	2.2	1.3	0.0	6.1
Aberdeen, SD	74.0	2.4	3.2	0.3	20.4	Aberdeen, SD	66.0	1.4	1.2	0.0	5.5
Livermore, CA	73.0	1.6	1.1	0.0	5.7	Monterey, CA	71.0	1.1	0.9	0.0	5.3
Monterey, CA	66.0	1.4	0.7	0.0	3.1	Livermore, CA	72.0	1.0	0.8	0.0	3.7
Yorktown, SK	76.0	0.8	0.7	0.0	4.3	Yorktown, SK	30.0	0.8	0.8	0.0	3.5
Penticton, BC	68.0	0.7	0.5	0.0	2.3	Penticton, BC	69.0	0.7	0.6	0.0	3.2

[a]For each season, the sampling sites are listed in order of highest mean concentration.

site in Vienna. Concentrations of SO_4^{2-} and acid aerosols in Ljubljana were comparable to those measured in this study, after accounting for the different sampling durations. Due to the limited number of European measurements available, we chose to compare our measurements primarily to the more extensive North American database, as well as other measurements collected in coal-burning areas (*10,27,28*) (Table 4).

Erfurt and Sokolov report aerosol acidity concentrations that are at the low end of the observations in North America and much lower than those measured in London during the 1963–1972 period (Tables 2 and 4). Winter PM_{10} and SO_2 concentrations at both eastern European sites were also much lower than those measured in London (*28*), but well above concentrations observed in the North American communities used for comparison (Tables 1, 3, and 4).

Mean concentrations of PM_{10} in Erfurt and Sokolov were significantly higher in the winter than in summer (Table 1), which is consistent with the occurrence of winter inversions and increased coal burning (particularly in Erfurt) for residential heating. Although the record of SO_2 sampling was not complete, concentrations also appeared to be higher in winter than in summer. During 1991, two consecutive 12-hr samples were collected daily in both Erfurt and Sokolov with the HEADS sampler. In Erfurt during February–April, the mean 24-hr SO_2 concentration was 125 μg/m^3 (SD = 122, range: 27–656, *N* = 35), while in May–September, the mean was 22 μg/m^3 (SD = 21, range: 3–93, *N* = 55). In Sokolov the February–April 24-hr mean SO_2 concentration was 76 μg/m^3 (SD = 45, range: below detection–205, *N* = 31), while in May–September, the mean was 31 μg/m^3 (SD = 22, range: 3–97, *N* = 46). Aerosol acidity was slightly higher in the summer than in winter at both sites (Table 2). This contrasts with observations from North America in which summer acidity levels are much greater than those measured in the winter (*3*) (Table 2).

Although acid aerosol levels were very low, comparisons to the North American database of SO_4^{2-} measurements (Table 2) indicates that the eastern European sites presented somewhat higher SO_4^{2-} concentrations, particularly during the winter period. As SO_4^{2-} concentrations can be viewed as "potential acidity," it is evident that little of the aerosol SO_4^{2-} was acidic, suggesting substantial neutralization. H^+/SO_4^{2-} ratios were below 0.4 for all samples except for one 24-hr winter sample collected in Erfurt (ratio = 1.25) and two winter samples collected in Sokolov (ratios = 0.60 and 0.65).

As suggested by the observations of low aerosol acidity and high SO_4^{2-} concentrations, levels of NH_3 were elevated (Table 4) in Sokolov and Erfurt. Ambient NH_3 measurements are indicative of the extent to which acid aerosols may be neutralized into nonacidic species, since NH_3 neutralizes acidic sulfate aerosols to produce neutral salt species. Although the record of NH_3 concentrations is less complete than

Table 4. Summary statistics of ambient concentrations (μg/m^3) measured in Sokolov and Erfurt and comparisons to measurements in Wuhan, China (*27*), Steubenville, Ohio (*10*) and historical measurements in London (*28*)

Site	Mean	SD	Peak
Aerosol acidity (as H_2SO_4)			
Sokolov	0.5	0.6	7.6
Erfurt	0.4	0.4	8.1
Wuhan	0.7	NA	2.4
Steubenville	1.2	NA	NA
London	6.5	6.7	134
SO_4^{2-}			
Sokolov	9.6	6.0	36.4
Erfurt	11.0	9.0	74.1
Wuhan	50.4	NA	94.1
Steubenville	12.8	NA	NA
NH_3			
Sokolov	2.6	3.2	23.6
Erfurt	1.7	1.1	13.9
SO_2			
Sokolov	52	63.7	592
Erfurt	60	94.5	715
Wuhan	42	NA	73
Steubenville	63	NA	NA
London	317	163	1298
Pariculate matter ≤10 μm (PM_{10})			
Sokolov	59	35	171
Erfurt	66	45	269
Wuhan	NA	NA	350
Steubenville	47	NA	NA
London[a]	92	71	709

NA, data not available from references.
[a]London PM_{10} measurements are estimated from British smoke measurements where British smoke is assumed to be equal to PM_{10} as a lower bound (*43*). Mean measurements of acidity and SO_4^{2-} from Erfurt and Sokolov are from Harvard impactor (24-hr) measurements; peak measurements for all species except PM_{10} are from HEADS (12-hr) measurements.

the aerosol acidity and sulfate measurements, mean concentrations during the sampling periods were consistently high (>1.4 μg/m^3) throughout the year. Annual averages in North American communities are typically below 1 μg/m^3, with only rural sites reporting annual means above 1.4 μg/m^3.

The observation of higher SO_4^{2-} levels in Wuhan, China (*27*) than in Erfurt or Sokolov is indicative of low conversion of SO_2 to SO_4^{2-}, given the higher SO_2 levels measured in Erfurt and Sokolov (Table 4). SO_2 levels in Erfurt and Sokolov were well below those measured in London, but significantly higher than those measured in China. In contrast, the Wuhan environment may have presented more complete conversion, but acidity appeared to be controlled by neutralization. In both the Wuhan study and this study, levels of aerosol acidity were surprisingly low, given the high measured concentrations of SO_2 and PM_{10}. PM_{10} concentrations in Erfurt and Sokolov were similar to concentrations measured in Wuhan. Although these comparisons are limited, they suggest that the conditions encountered in Erfurt and Sokolov may be typical of regions in which high-sulfur coal is burned during the winter. In such situations, although SO_2 levels may be greater than concentrations observed in North America, aerosol acidity levels are very low.

Sampling and Analytical Issues

One explanation for our inability to observe aerosol acidity lies with the measurement itself. Were the measurements used in this study unable to measure acidity that was present in the atmosphere, or were the studies in London in the 1950s and afterwards measuring acidity produced via artifact reactions? Although few data are presented, the analytical method used in London was apparently insensitive to artifact formation of acid from SO_2, even though only a small amount of SO_2 would need to react on the filter to produce a relatively large artifact of sulfate (*29*). At the levels of SO_2 measured in London (Table 4), this artifact could easily account for all of the measured aerosol acidity. In future studies in similar environments it would be advisable to attempt to directly replicate the measurement method of Commins (*29*).

Although there was no measured acidity, we examined the possibility that the high particle concentrations led to particle–particle interactions on the filter which resulted in an apparent loss of measurable acidity. For this analysis we used the data (approximately 230 samples) from the multistage filter pack of the HEADS (Table 5). The acid correction is minimal, although there is a tendency for a negative correction due to higher NH_4^+ on the backup filter. These results are consistent with those reported by Koutrakis et al., who found little correction for samples collected in six locations in North America (*22*). Although NH_4^+ concentrations were quite high, most of the NH_4^+ and NO_3^- was found on the Teflon filter and not on the backup, suggesting only minimal volatilization of NH_4NO_3. Ion balances ([H^+] + [NH_4]/2 ([SO_4^{2-}] + [NO_3]) on the Teflon filter were only slightly less than 1, suggesting that all anions associated with acidity were accounted for (Table 5).

Alternatively, although the method used in the measurements reported here is suitable for generated atmospheres of acidic sulfate species (*30*) and for acidic aerosols produced during summer photochemical processes (*18*), it may not adequately measure aerosol acidity that is likely produced via heterogeneous mechanisms and associated with carbon particles. This question remains unanswered because all applications of this method in winter pollution situations (*7,27*), including our study, have measured little or no acidity, while the method has only been evaluated for aerosols likely to resemble those produced in summer conditions.

We also investigated the possibility that in the humid environments with high concentrations of particles characteristic of winter episodes, acidity was present in larger size fractions than collected by our sampler. Measurements of SO_4^{2-} in the PM_{10} fraction were made on a subset of the samples collected in Erfurt (N = 35) and Sokolov (N = 45). SO_4^{2-} in the PM_{10} and $PM_{2.5}$ fraction were highly correlated (r^2 = 0.94), and there was little indication of additional SO_4^{2-} in the PM_{10} fraction. This limited analysis suggests that no additional aerosol acidity would be associated with particles of aerodynamic diameter >2.5 μm.

Acid Production and Neutralization

Another explanation for our observation of low aerosol strong acidity is the effect of low SO_4^{2-} production or neutralization of acidic sulfate species by ambient NH_3. The meteorological conditions of local winter inversions suggests that SO_2 and consequently SO_4^{2-} ("potential acidity") concentrations and NH_3 will peak at the same times, limiting the opportunity for acidic particles to avoid neutralization. This contrasts with North American summer episodes, for which it is believed that convective mixing replaces stagnant surface-level air (high ammonia content) with acid-laden air that has been transported above inversion layers where it is protected from neutralization (*3,31*). The situation in eastern Europe also differs in terms of the proximity of the sources, with local sources emitting SO_2 below inversion layer heights and therefore in close proximity to NH_3 sources, providing ample opportunity for neutralization.

Further, the SO_2 conversion reactions are expected to differ between the (eastern European) winter and the North American summer, where photochemical reactions predominate. In North America, emissions are typically above the height of inversion layers under conditions of low particle concentrations, facilitating transport of gaseous species at heights where slow conversion may occur while the air mass is protected from neutralization by surface level sources of NH_3. The predominant conversion mechanism in North America is photochemical, based on the reaction of hydroxyl radical with SO_2 in the presence of water (*32*). In eastern Europe, where winter inversions reduce the impact of photochemistry and where high particulate concentrations provide sufficient surface area, heterogeneous reactions are expected to predominate.

Aqueous-phase oxidation mechanisms may explain our observations of lower than expected sulfate levels at a given SO_2 concentration, as well as the high levels of NH_4^+ ion observed (mean concentrations >4 μg/m^3). In the winter inversion setting, concentrations of oxidizing species may be quite low and conversion of SO_2 limited. Homogeneous gas-phase conversions, associated with North American summertime acidity, occur at rates of 0.3–2%/hr, while aqueous phase (nonmetal catalyzed) oxidation is slower (0.2%/hr) (*33*).

The extent of SO_2 conversion may be estimated by the [SO_4^{2-}/SO_4^{2-} + SO_2] ratio. Much like the North American situation, the ratios in Erfurt and Sokolov were lower in the winter than in the summer. Ratios

Table 5. Summary statistics for denuder/filter pack measurements of particulate species[a]

Site	H^+ uncorrected	H^+ correction	H^+ total	Ion balance	SO_4^{2-} (teflon)	NO^{3-} (teflon)	NH_4^+ (teflon)	NO_3^- (backup)	NH_4^+ (background)
Sokolov	13.6	-8.4	15.9	0.95	99.3	51.9	225.0	8.1	14.6
Erfurt	8.9	-19.3	12.9	0.89	109.7	48.8	245.6	14.6	37.8

[a]All concentrations in nmol/m^3. All values are mean values for the (approximately) 230 denuder samples collected (i.e., the H^+ total is the mean of the 230 H^+ total measurements, not the sum of the mean H^+ uncorrected + mean H^+ correction). H^+ correction refers to F2 NO_3^- – F4 NH_4^+. H^+ total = H^+ uncorrected + H^+ corrected.

were typically below 0.25 in the winter, and reached peaks of 0.6 or higher in the summer. While these ratios suggest considerable conversion in the summer, SO_2 concentrations were low during this period. In the winter, when SO_2 concentrations are elevated, $[SO_4^{2-}/SO_4^{2-} + SO_2]$ ratios were below mean ratios of 0.4–0.6 seen in summer acidic atmospheres in North America (*21,31*), suggesting the impact of slower SO_4^{2-} production processes.

Implications

Analysis of aerosol acidity as a causal factor distinct from fine particulate matter is particularly important for North America because levels of fine particulates may be elevated throughout the year and in regions without high acid aerosol levels. In contrast, aerosol acidity is elevated during the summer, and high concentrations are usually only found in the eastern portion of the continent (*3*). The comparisons presented in this article suggest that while aerosol acidity may be present in elevated concentrations during summertime episodes in eastern North America, there are few other settings with similar levels of aerosol acidity. In particular, areas where high-sulfur coal was burned showed low levels of acid aerosols. Additionally, short-term acidity concentrations typically measured in North America are well below levels shown to elicit effects in controlled chamber experiments (*3*). These findings when viewed along with the observation that in summer episodes in North America acid particles are inherently correlated with fine particulate levels, makes it difficult to distinguish the acidic fraction as a causal agent. Therefore, analysis of the health impact of inhalable particulates in settings with elevated particle concentrations (*5–8,14,34,35*) where acidity is low suggests that aerosol acidity may not be the primary particulate parameter associated with increased morbidity and mortality.

Further evidence for the absence of aerosol acidity as a causal factor in air pollution health studies comes from its low indoor:outdoor ratio (*12,13*). Although fine particles penetrate efficiently indoors so that indoor:outdoor ratios are approximately 0.8 (*36,37*), acid levels are quite low indoors as a result of indoor ammonia, which neutralizes aerosol acidity. Indoor:outdoor ratios of sulfate are also near 0.8 (*12,38*). Accordingly, outdoor concentrations of fine particles and sulfates are likely to be better indicators of total exposure than are acid aerosols because their indoor:outdoor ratios are high and since the majority of an individual's time is spent indoors.

Much of the epidemiological evidence also does not support aerosol acidity as the primary causal factor in the health effects of particulate air pollution. In addition to the comparative analysis of St. Louis and Kingston-Harriman Tennessee (*9*), Dockery and colleagues analyzed the Harvard Six-Cities Study and reported that mortality was more strongly associated with levels of fine and inhalable particulates, as well as sulfate particles, than it was with aerosol acidity (*10*). One of the cities included in this analysis, Steubenville, Ohio, had air quality levels which were quite similar to those measured in this study (Table 4). These data, along with the Utah Valley (*5–8*) and Seattle (*35*) studies, and results presented here indicate either stronger associations between health effects and PM_{10} mass than aerosol acidity or relationships between particle pollution and health in areas where aerosol acidity levels are low.

In an investigation of hospital admissions in Toronto, Thurston and colleagues (*11*) found a stronger relationship with aerosol acidity than with either fine particles or inhalable particles; however, this result is not in direct conflict with studies that show weaker relationships with aerosol acidity than with other particle metrics. As Thurston et al. have discussed (*11*), aerosol composition may differ dramatically even in locales where significant associations were observed between inhalable particulates and health effects. In the Toronto area, during summer episodes the submicrometer aerosol is dominated by acidic sulfate aerosols. It is plausible that in this setting aerosol acidity merely acts as a surrogate for the submicrometer aerosol which most fully penetrates into indoor environments. In regions such as Erfurt and Sokolov where submicrometer aerosols are not acidic, other particle metrics may be associated with health effects. Future epidemiological studies should collect more detailed information on size-fractionated particle composition, with special emphasis placed on the $PM_{2.5}$ fraction which penetrates indoors.

As Dockery and Pope have suggested (*39*), the available epidemiological data indicate that the associations observed between particulate air pollution and health effects are due to the mass concentration of the particle mix common to urban areas rather than to specific chemical species within the mix. Even in locales where particle composition, as it is traditionally measured, is quite different, and where major particle sources are different (auto exhaust, woodsmoke, steel mill emissions, transported power plant emissions), relationships between particle concentrations and measured health outcomes appear to be remarkably consistent. One common feature of the particulates in these studies is that they are produced in combustion processes. Studies of naturally produced particles show a much smaller impact on health outcomes for a given particle concentration (*40*). These data support the hypothesis that any particulate air pollution produced by combustion will be associated with adverse health outcomes. The implication of this hypothesis is that particulates are associated with adverse health effects in all settings with combustion air pollution, or essentially all urban areas.

Although the identification of specific inhalable particle components associated with adverse health outcomes is important for understanding air pollution epidemiology and for implementing control strategies, perhaps the most compelling reason to investigate the relationship between particle composition and health outcomes is to understand the biological mechanism by which airborne particulates may cause effects. The epidemiological evidence which suggests that aerosol acidity is not the primary particle parameter associated with health effects implies that the biological mechanism associated with particulate air pollution may be different from that experienced in animal and controlled chamber studies with sulfuric acid aerosol exposures. That adverse health effects may be found in settings where aerosol acidity is not observed indicates that other particle parameters in addition to aerosol acidity should be investigated in both epidemiological and mechanistic studies. For example, recent evidence from *in vitro* studies suggests that iron present on the surface of particles may promote lung injury (*41,42*). For epidemiologists it is important to identify settings with different particle compositions and to investigate whether the health effects are still observed. In particular, thorough analysis of particle composition for PM_{10} samples collected during routine monitoring is recommended, especially for locations where particle-associated health effects have already been observed.

Conclusions

A large number of studies have indicated associations between particulate air pollution and adverse health outcomes. Wintertime air pollution in particular has been associated with increased mortality. Identification of causal constituents of inhalable particulate matter has been elusive, although one candidate has been the acidity of the aerosol. Here we report on low levels of aerosol acidity in relatively polluted environments which were directly impacted by the burning of high-sulfur coal. These measurements, along with reported associations between fine particulate air pollution and health outcomes in other regions where little aerosol acidity has been measured, suggest that particulate acidity alone is not the primary component defining the toxicity of fine particulate air pollution.

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